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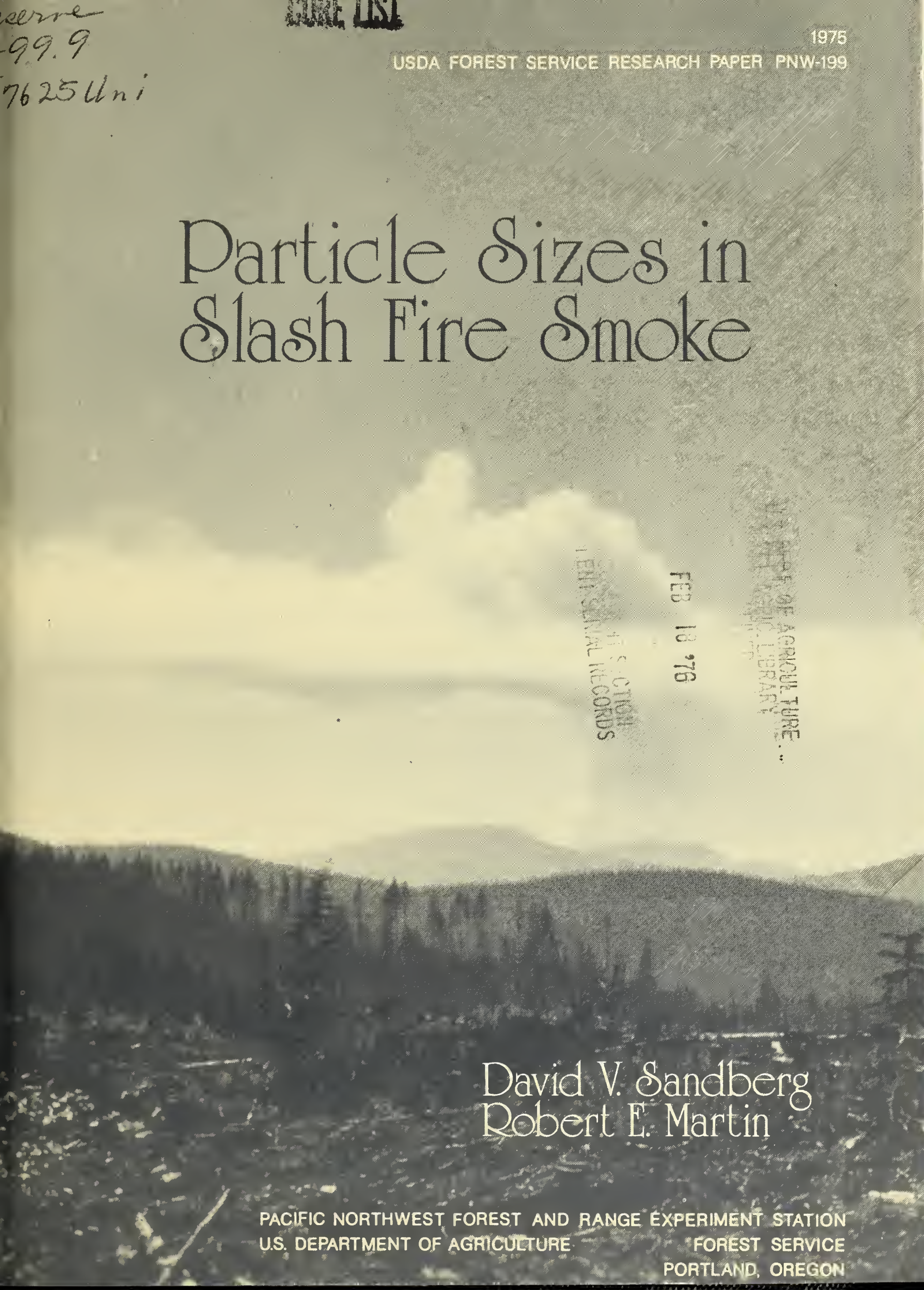

Particle Sizes in Slash Fire Smoke

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PARTICLE SIZES IN SLASH FIRE SMOKE

Reference Abstract

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Particulate emissions are the most objectionable atmospheric contaminant from forest burning. Little is known of the particulate sizes, and this research was done under laboratory conditions to obtain particle size information. Comments are made concerning techniques for future work in this field.

KEYWORDS: Air pollution (slash disposal), prescribed burning (-particulates.

RESEARCH SUMMARY
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Forest burning contributes a substantial amount of emissions to the atmosphere each year. The particulate fraction of the emissions is most objectionable from the standpoint of visibility, esthetics, and atmospheric degradation. Little is known of the sizes and configuration of the particulates, and this study was conducted to gain a better understanding of particulate characteristics emitted in slash burning. Impactor studies revealed 82 percent of particulate mass was less than 1-μm (micrometer) aerodynamic diameter and 69 percent was less than 0.3 μm. Scanning electron microscopy revealed four types of particles: (1) single spherical submicrometer particles 0.05- to 0.6-μm diameter with most of them close to 0.1 μm; (2) an agglomeration of the first particle type into chains up to 4 μm long or spheres up to 80-μm diameter and containing at least 200 small particles; (3) nearly spherical particles from <1-μm to 12-μm diameter; (4) angular particles from <0.05 to about 20 μm in largest dimension.

Prescribed forest burning is a source of emissions into the atmosphere in the Pacific Northwest, as it is in some other forested regions in the United States. Burning of forest fuels has been estimated to produce 23.7 percent of the 6.1 million metric tons of particulates emitted to the atmosphere each year and 6.9 percent of the 2.0 million metric tons of hydrocarbons (National Air Pollution Control Administration 1970)(a metric ton equals 1.12 short tons). Increased public awareness of air pollution has focused unfavorable attention on the burning of logging slash. The burning of 6.3 million metric tons of forest residue in Washington and Oregon annually releases 45,000 metric tons of hydrocarbons and 76,000 metric tons of particulate matter into the atmosphere.[1/] Wildfires contribute even more, but estimates are difficult to make.

The purpose of this paper is to present data on the relative mass of submicrometer (<1-μm or $<10^{-6}$-m), large (1- to 5-μm), and giant (>5-μm) particles

[1/] G. R. Fahnestock. A problem analysis--impacts of forest residues and their disposal on forest land management and environment in the Pacific Northwest. Unpublished report, 28 p., 1968. On file, Pacific Northwest Forest and Range Experiment Station, Portland, Oreg.

released from the laboratory burning of slash-type fuels. In addition, the size range and shape of particles will be reported. In part, this was a pilot study, and we'll comment on appropriate techniques for future slash burning air pollution studies.

PREVIOUS WORK

First, we might delineate some general air pollution problems that are not created by forest burning. Sulfur dioxide is not created by forest burning, and temperatures are too low for the emission of significant amounts of the oxides of nitrogen. The hydrocarbons formed are generally not photochemically reactive, nor have they been shown to be a health hazard. The worst that can be said of them is that they may condense to form visible aerosols (Hall 1972).

Particulate emissions, on the other hand, can degrade air quality by reducing visibility. Northwest air is polluted more by particulates than other pollutants when compared by Federal standards,[2/] so reduction of particulate emissions in smoke sensitive areas is the current priority of most local air pollution authorities. Also, there is a generally negative public reaction to any smoke or other material that is added to the atmosphere, whether or not it constitutes air pollution (Cramer 1969).

Little is known directly about the quantity or quality of pollutants released from burning of forest residues; most of what is known is extrapolated from experiments of burning other agricultural fuels. Many components of the hydrocarbon fraction have been described (Fritschen et al. 1970), and other relevant experiments are underway. Hall (1972) summarizes the knowledge. Of the important particulate fraction, however, the very least is known.

Of all the properties of particles in the air, none dominates their behavior more (nor is more difficult to study experimentally) than size. The laws governing many important properties of aerosols, including light scattering, coagulation, predominance of diffusion or settling, rate of cooling, and microscopic visibility, change fundamentally as the particle radius approaches the mean free path of the gas molecules, 0.07 μm in air at standard conditions; or with the average wavelength of visible light, 0.55 μm. The effect of an aerosol on visibility is determined more by particle size than by concentration. In general, a particle of 1 μm or more in diameter scatters light in proportion to the square of its radius; a submicrometer particle scatters light in proportion to the sixth power of its radius (Fuchs 1964).

The mechanisms for removal of particles from the atmosphere also depend on particle size. In general, submicrometer particles have settling velocities less than 3.5×10^{-3} $cm \cdot s^{-1}$, are dominated by Brownian motion, diffuse in air, and remain suspended for days or more, while larger particles will settle out of still air. Particles with a radius comparable to or smaller than the mean free path of air molecules will not settle out (Green and Lane 1964).

Numerous authors, as summarized by Brown et al. (1950), have recognized the relationship between particle size and lung penetration in man. They conclude that nasal efficiency for screening out airborne particles entering the respiratory tract is 100 percent for particles above 5 μm and decreases with size to zero for 1-μm particles. They also found that alveolar retention is complete for particles larger than 1 micrometer that are not filtered out by the upper respiratory tract. Optimum particle size with the highest probability of alveolar deposition is about 1-μm diameter. Smaller particles down to 0.25 μm have about the same probability of being retained in the alveoli as those from 1 to 5 μm. Wells (1955) pointed out that it is not the projected diameter but the aerodynamic dimension that determines lung penetrability.

[2/] Washington State Department of Ecology. Summary of the implementation plan for attainment of the national ambient air quality standards. Mimeographed, 30 p., 1972.

Aerodynamic dimension is the size of a spherical particle that would have the same falling velocity as the particle described. Projected diameter is the diameter of a circle having the same area as a cross section of the particle.

There is some controversy in the literature concerning the particle sizes in wood fire smoke. Feldstein and his associates (1963) estimated that 50 percent of the particulate mass from the burning of land clearing debris is in the 50- to 100-μm size range. The data were derived from the examination of microscope slides placed around fires to collect fallout. However, Meland and Boubel (1966) measured smoke from field burning and found that particles were uniformly distributed in the submicrometer range. Haessler (1965) said that smoke particles, as initially formed, are usually about 0.1 μm. MacArthur (1966) collected particles from bush-fire smoke in a thermal precipitator. They ranged in size from 0.1 to 1.0 μm, with a marked preponderance at about 0.1-μm diameter. Vines and others (1971) conducted extensive experiments. Using a variety of sizing techniques and aircraft traverses of smoke plumes, they found most of the particles collected appeared to be close to 0.1-μm diameter, and that smoke contained few particles larger than 5 μm. Some agglomerates of smaller particles were found as large as 50 μm. Two types of small particles were found. Spherical, smooth particles thought to be tar were 0.2 μm or more in diameter. Rough, crystalline ash and carbon particles between 0.05 and 0.03 μm were found to be less numerous than the tar particles. Butcher and Charlson (1972) report that condensation of particles in flames produce submicrometer particles that may coalesce or agglomerate to clumps or chains before being emitted from the source.

Particulates may be the most important pollutant emitted from forest burning, but the least is known about them. Aerodynamic particle size is a most important characteristic of particles, but there is disagreement on the proportion of large particles in smoke. Particle size determines the residence time of the particles in the air and their penetrability into human lungs. Large particles scatter light inefficiently, fall out rapidly, and are less important air pollutants than submicrometer particles.

METHODS

Douglas-fir (*Pseudotsuga menziesii* (Mirb.) Franco) slash was collected from a second-growth stand in western Washington 1 year after harvesting. From branches and twigs, a continuum of sizes less than 3-in (7.6-cm) diameter, 17 fuel beds were constructed in the laboratory. The surface-to-volume ratio of each fuel bed was 86 ft^2/ft^3 (282 m^2/m^3). Porosity, the ratio of fuel bed volume to fuel particle volume, ranged from 9 to 18 at ignition. Fuel moistures ranged from 11 to 23 percent of ovendry fuel weight. The fuel beds weighed 13 lb (5.9 kg), corresponding to a slash loading of about 38 tons/acre (84 metric tons/ha).

The circumference of the fuel beds was ignited with a propane torch. Fuel weight loss was continuously recorded and converted to energy release rate in Btu $ft^{-2}\cdot min^{-1}$ ($k\ cal\cdot m^{-2}\cdot min^{-1}$ and $J\cdot m^{-2}\cdot min^{-1}$). Smoke from the fires was contained in a hood vented by a 16-in (40.6-cm) diameter constant-volume-flow induced-draft flue. A sample was removed 15 ft (4.6 m) from the fuel bed via a 25-in-long (63.5-cm) sample line with a sharp inlet nozzle of 3/8-in (0.95-cm) diameter. The nozzle size provided isokinetic sampling within 10 percent, using a constant sample volume flow of 1 $ft^3\cdot min^{-1}$ (0.0283 $m^3\cdot min^{-1}$) at standard conditions.

The samples were drawn through an eight-stage Andersen impactor (Andersen 1966).[3/] According to the manufacturer,

[3/] Mention of products by name does not constitute an endorsement by the U.S. Department of Agriculture.

the first two stages remove nearly all material over 5-μm aerodynamic diameter, three stages remove large material, and two stages remove submicrometer particles to about 0.3 μm. A 47-mm inline Gelman Type A glass-fiber filter as the last stage removes nearly all remaining solids or condensed liquids. Temperatures at the sample line inlet never exceeded 200° F (93° C). That temperature in the impactor produces only a 10-percent error in the indicated diameter of particles on each stage. The seven stainless steel impactor stages and the filter mat were weighed before and after sampling. The collection plates were cleaned only after five fires were sampled to allow adequate accumulation for accurate weight measurement.

Smoke from four of the fuel beds was sampled during the smoldering (postflaming) period only. The average intensity of the four fires was 126 Btu $ft^{-2} \cdot min^{-1}$ (342 k $cal \cdot m^{-2} \cdot min^{-1}$ or 1.43×10^4 $J \cdot m^{-2} \cdot min^{-1}$). Five fires, with intensities ranging from 353 to 1,417 Btu $ft^{-2} \cdot min^{-1}$ (958 to 3845 k $cal \cdot min^{-2} \cdot min^{-1}$ or 4.01×10^4 to 16.1×10^4 $J \cdot m^{-2} \cdot min^{-1}$), were sampled during the entire flaming period. Eight fires were sampled only while the intensity was greater than one-half the peak for that fire. Intensities for that series ranged from 1,000 to 1,857 Btu $ft^{-2} \cdot min^{-1}$ (2712 to 5040 k $cal \cdot m^{-2} \cdot min^{-1}$ or 11.4×10^4 to 21.1×10^4 $J \cdot m^{-2} \cdot min^{-1}$).

Five additional fuel beds were burned to obtain samples of 0.4-μm pore diameter nucleopore filter media for electron photomicrographs. Thirty-four samples, representing all phases of fire activity, were collected. One filter was placed on each stage of the Andersen sampler. Oblique photographs, with a 30-degree tilt angle, were taken on a scanning electron microscope. The photographs were used to determine the shape and size range of the particles.

RESULTS

The electron microscope revealed four distinct types of particles. By far the most prevalent was the singular spherical submicrometer particle reported by other investigators (fig. 1). The

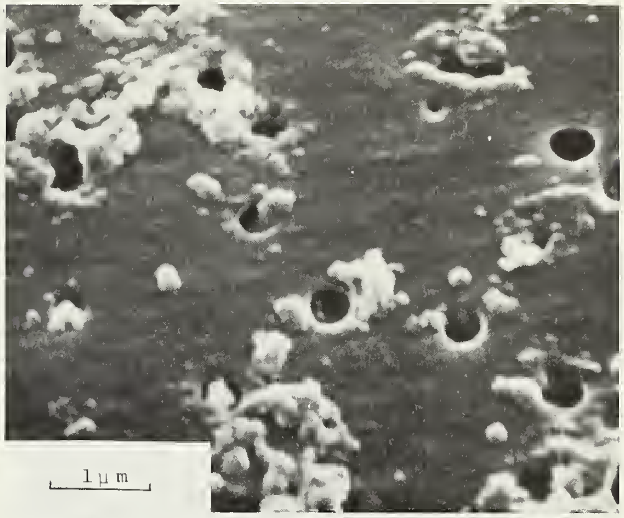

Figure 1.--Spherical submicrometer smoke particles from burning Douglas-fir slash (Scanning Electron Micrograph-SEM).

observed range of projected diameters was from less than 0.05 to 0.6 μm. The limit of resolution in the photographs was slightly less than 0.05 μm. Most of these spherical particles were very close to 0.1 μm. These particles adhered to filter media, and solvents were required to remove them from metal surfaces.

The second type of particle observed was an agglomeration of the spherical particle above. Branched chains of particles up to 4 μm long and containing at least 200 submicrometer spheres were seen, but not commonly (fig. 2). Roughly spherical agglomerates of a few to an estimated 10^8 small particles were commonly observed (fig. 3). The largest cluster seen had a projected diameter of about 80 μm (not shown). The agglomerates accounted for the vast majority of all particles present with aerodynamic

diameters over 1 μm. Agglomeration was apparently enhanced by the walls of the sampling line. When a 6-ft-long (1.83-m), 1/4-in-diameter (0.64-cm) sampling line was used, nearly all submicrometer particles agglomerated to large clusters. Agglomeration was still very common when a short (4-in or 10.16-cm), large-diameter (5/8-in or 1.59-cm) sample line was used, however.

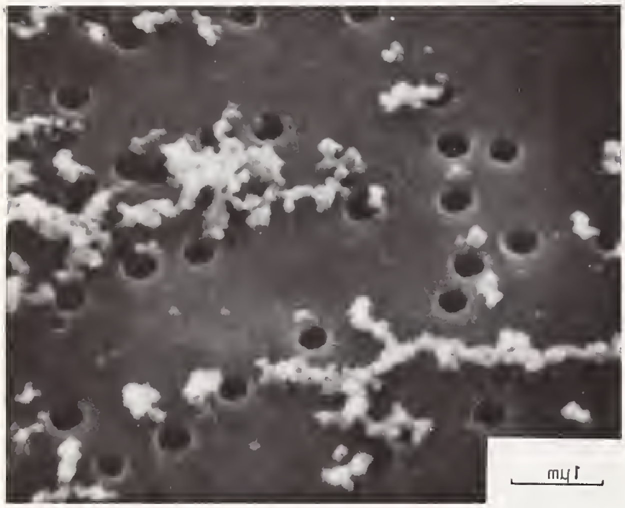

Figure 2.--Agglomerated chain of smoke particles from burning Douglas-fir slash (SEM).

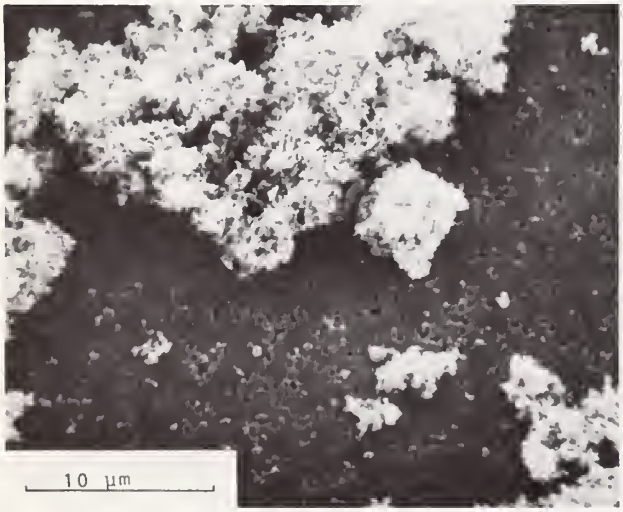

Figure 3.--Large clusters of spherical submicrometer smoke particles from burning Douglas-fir slash (SEM).

Solid, nearly spherical particles were also seen, ranging from less than 1 μm to 12 μm (fig. 4). The surface of

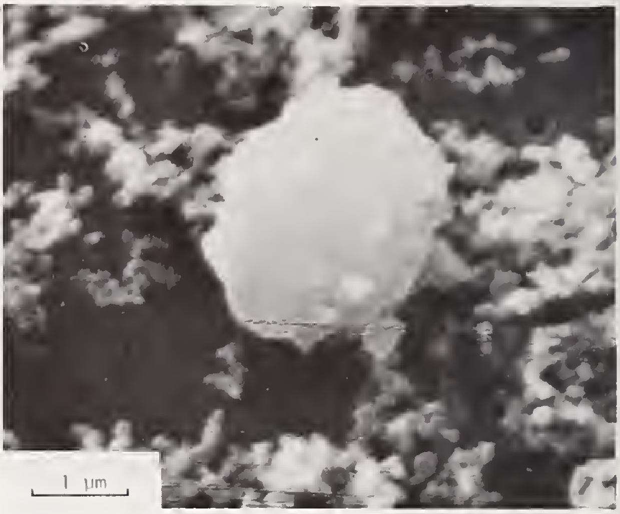

Figure 4.--Large, smooth, solid smoke particle from burning Douglas-fir slash (SEM).

the particles was textured with round protrusions of less than 1-μm diameter, suggesting that they are also agglomerates of the submicrometer spheres above. These particles were rare and, in this study, were never observed from a smoldering fuel bed.

Finally, some solid, angular particles were seen in the photographs. These ranged in size from less than 0.05 to about 20 μm. These particles were very common from smoking fuel that had not yet produced flames (fig. 5) but were present in lesser quantities during all phases of fire activity. These particles did not adhere to filter media or to each other. Occasionally, a giant particle of this type did not remain on the impactor plate where it had been collected; this caused a small underestimation of the number of giant particles.

Comparison of photographs revealed that the preflaming smoking fuel bed released a high proportion of giant particles. The smoldering phase appeared

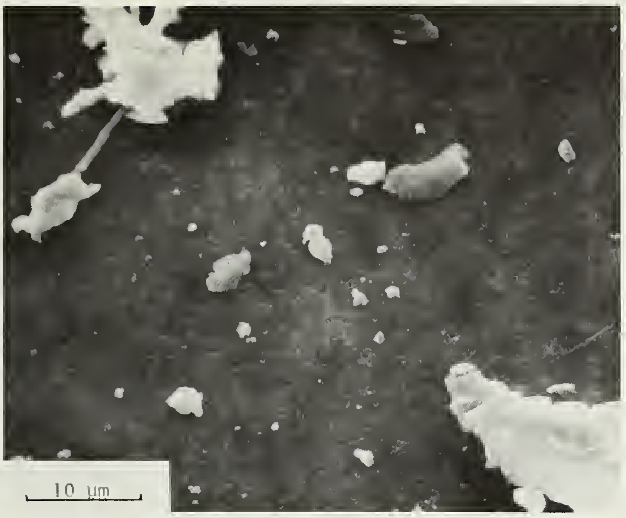

Figure 5.--Solid, angular smoke particles from preignition phase of burning Douglas-fir slash (SEM).

to have the highest proportion of submicrometer particles, but no particle count could be made.

The impactor was used to separate smoke particles at 5-, 1-, and 0.3-µm aerodynamic diameters. The relationship of particle mass to fuel weight loss for each size class was calculated. The coefficients of variation between fires within series were high, from 0.30 to 1.43. The series of fires for which the flaming portion was sampled released 82 percent of the particulate mass in the submicrometer range. The smoldering period submicrometer fraction represented 87 percent of the mass. However, the variation was too high to make a statistically valid comparison between the emission behavior of different periods of the fire, or different intensities of burning.

The data from all 17 fires were pooled and weighted by the relative amount of fuel consumed by each. It was found that 8 percent of the total particulate mass is larger than 5-µm aerodynamic diameter; 10 percent is between 1 and 5-µm; 13 percent is between 0.3- and 1-µm; and 69 percent is smaller than 0.3-µm.

SOURCES OF ERROR

Several possible sources of error were present in this experiment. The fuel bed included only small limbs and twigs, to the exclusion of the large chunks, stumps, live fuel, needles, rotten logs, and forest floor present in any field burn. The interaction between wind and flames that causes erratic fire behavior on field burns was absent in the laboratory. It is possible that the higher local fire intensity, more powerful fluid dynamic forces, and the varied fuel bed composition in a field burn might increase the fraction of giant particles emitted.

The Andersen sampler used was designed as an ambient air sampler but was used here as a source sampler. The addition of an inlet sampling line probably increased the agglomeration of particles. Some impaction of giant particles occurred on the first jet stage of the sampler. This impaction systematically removed a small fraction of the very large (20-µm) particles.

Occasionally, a partially consumed needle, wood sliver, or bark fragment could be entrained in the plume and deposited on a filter. These particles would constitute only a fraction of 1 percent of the particulate matter sampled. Such particles would fall out very quickly. By chance, none were included on any of the impactor plates in this study, and their existence is mentioned here only as a possible source of error in future studies.

The weight of smoke particles in one size class collected from one fire was frequently in tenths of milligrams. Weighing errors could be as large as 20 percent in these cases. This was avoided somewhat by the accumulation of the smoke particles from several fires on one plate. Some slippage of the largest particles through the first collection plates occurred, allowing a small underestimation of the giant particles.

Errors due to non-isokinetic sampling and leakage and those caused by not compensating for temperature are thought to have been held to less than 5 percent.

DISCUSSION

Electron photomicroscopy of particulate emissions from the laboratory burning of logging residues confirmed the existence of two basic particle structures. The most abundant particle was a smooth, spherical, submicron particle suspected to be condensed tars. These spherical particles ranged from smaller than 0.05 μm to agglomerates of 80 μm. There was a marked preponderance of particles of about 0.1-μm diameter. The second basic particle group was polydisperse in size from infinitesimal to at least 20 μm and did not tend to agglomerate.

Impactor separation of particles reveals that 82 percent of the particulate mass was in the submicrometer range, 69 percent with an aerodynamic diameter less than 0.3 μm. The size distribution did not depend on fire intensity in the range sampled, as long as flaming existed. Smoldering fuel beds seemed to produce a higher percentage of submicrometer particles, but this was not demonstrated statistically. Samples collected from a smoking fuel bed before flaming started consisted almost entirely of giant, solid, angular particles.

An Andersen sampler designed for describing particle size distribution in ambient air was found to be satisfactory but imperfect for source sampling. However, the sample inlet eliminated some giant particles and enhanced agglomeration.

In experiments to determine emission factors from forest fuels, isokinetic sampling is probably not a critical requirement due to the rarity of giant particles. Isokinetic sampling is generally thought to be important for particles larger than 3-μm aerodynamic diameter.

The research reported herein measured particles only a few feet above the source. Additional sampling farther downstream might show a different distribution as hydrocarbons continued to condense, and particles agglomerated. A companion study could be accomplished by aerial sampling downwind from field burns, or within a duct system in the laboratory.

A multistage impactor seems to be a good tool for describing an aerosol with the wide range of sizes in slash fire smoke. However, a sampler specifically designed for source sampling would be better. At any rate, a separate sampler should be used to describe particles larger than 20 μm. The sample line should be as short and as large in diameter as practical, to reduce agglomeration.

A dense emission of smoke just before flaming occurs in forest fuels is common. Further work with electron microscopy is required to confidently describe this aerosol, which seems to be unique.

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